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Controllable synthesis and property of graphene-based magnetic metal nanostructures

Kong-Lin Wu [a], Xiang-Zi Li [b], Xian-Wen Wei [a, *], Ting-Hui Ding [a], Miao Jiang [a], Wen-Juan Zhang [a], Yin Ye [a]

[a] College of Chemistry and Materials Science, Key Laboratory of Functional Molecular Solids, The Ministry of Education, Anhui Laboratory of Molecular-Based Materials, Anhui Normal University, Wuhu 241000, PR China
[b] Department of Chemistry, Wannan Medical College, Wuhu 241002, PR China

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ABSTRACT

A facile and effective solution phase reduction method was developed to synthesize graphene-based magnetic metal nanocomposites. Metals (Co, and Ni) or alloys ($Fe_{51}Co_{49}$, $Fe_{48}Ni_{52}$, $Ni_{49}Co_{51}$, $Co_{51}Cu_{49}$, and $Ni_{52}Cu_{48}$)/reduced graphene oxide (RGO) nanocomposites were successfully prepared by reduction of the corresponding aqueous metal ions and ethylenediamine (EDA)–graphene oxide (GO) with hydrazine hydrate at 353 K for 1 h under N_2 atmosphere. The effects of synthetic parameters such as metal ions concentration, adding sequence of NaOH and $N_2H_4 \cdot H_2O$, linkage agent and reaction time on the formation of nanocomposites were investigated. The experimental results showed that using ethylenediamine and adding sequence played critical roles in the formation of metals or alloys/RGO nanocomposites. Magnetic hysteresis measurements revealed that the as-synthesized metals or alloys in nanocomposites showed excellent soft magnetic behavior with enhanced saturation magnetization, and could have promising applications in biotechnology, catalysis, and magnetic storage devices.

1. Introduction

Graphene, as the two-dimensional monolayer form of sp^2-hybridized carbon, has attracted research attention intensively due to its high surface area and superexcellent thermal, electrical, mechanical, and chemical properties [1–6]. It can also be served as an ideal support material with improved interfacial contact and enhanced properties [5,6]. Recently, the graphene/graphite-based magnetic metals or alloys composites have been investigated [7–19], since they had potential applications in fields of magnetic resonance imaging [8–11], microwave absorbing materials [13], and catalysis [18,19], *etc.* Simultaneously, the different synthesis methods such as chemical vapor deposition (CVD) method [8–11], a carbon-deficient CVD process [12], a co-deposition and subsequent annealing process [13], the CO Boudouard reaction [14], a high temperature annealing process under vacuum [15], and pyrolysising an organic metal/graphene composite precursor [16], were developed to prepare FeCo [8–11,13], NiCo, FeNi [12], Fe_2Co, $Fe_{0.64}Ni_{0.36}$ [14], and Co [14–16] -based graphene/graphite nanocomposites. Recently, a wet chemical reduction method [17–21] was used to fabricate reduced graphene oxide/Ni (RGO/Ni) nanocomposites [17,21], Ni_xCo_{1-x} alloy, Fe_xNi_{1-x} alloy, and Co nanoparticles/RGO nanosheets [18–20] by reduction of the corresponding metals ions and graphene oxide with hydrazine at more than 373 K [17–20] or under hydrothermal condition [21]. However these synthetic approaches need relative high temperature or rigorous reaction conditions. Therefore, developing a facile and mild strategy for preparation of graphene/graphite-based magnetic metals and alloys nanocomposites is still a challenge.

In this work, magnetic metals (Co and Ni) or alloys (FeCo, FeNi, NiCo, CoCu, and NiCu)/RGO composites were successfully prepared by a facile and effective solution phase reduction route in water at a

* Corresponding author. College of Chemistry and Materials Science, Key Laboratory of Functional Molecular Solids, The Ministry of Education, Anhui Laboratory of Molecular-Based Materials, Anhui Normal University, No.1 East Beijing Road, Wuhu 241000, PR China.
E-mail address: xwwei@mail.ahnu.edu.cn (X.-W. Wei).

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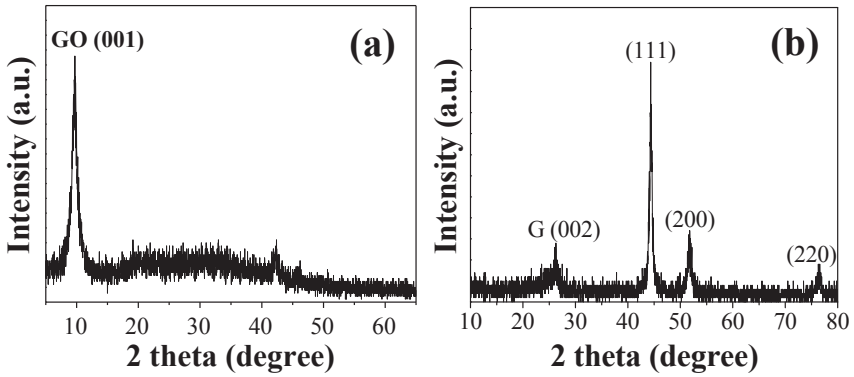

Fig. 1. XRD patterns of (a) GO and (b) Ni/RGO-1.

relative low temperature for 1 h under N_2 atmosphere. The effects of experimental parameters such as initial metal ions concentration, adding sequence of NaOH and $N_2H_4 \cdot H_2O$, linkage agent and reaction time on the formation of nanocomposites were systematically studied. It is important to use ethylenediamine as linkage and choose adding sequence of NaOH and $N_2H_4 \cdot H_2O$. The dependence of magnetic properties on the metal content in a series of metals or alloys/RGO composites had been explored. The as-

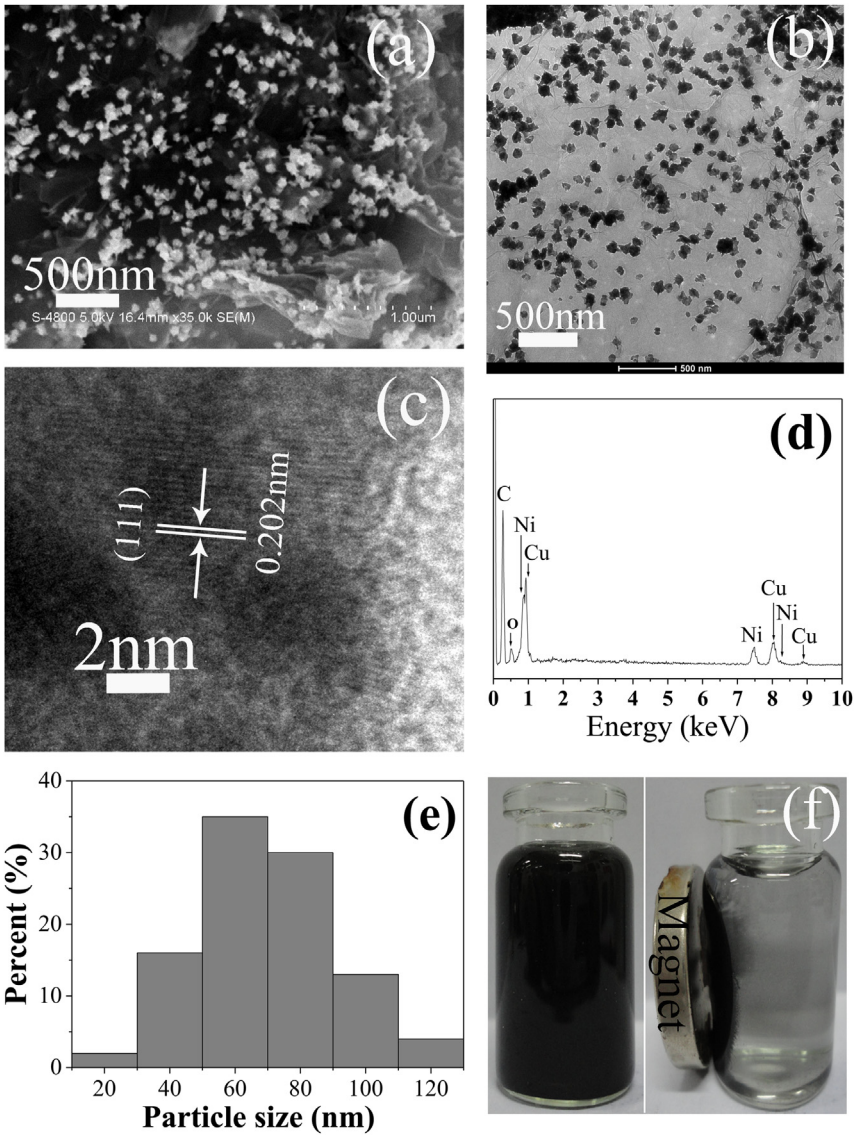

Fig. 2. Integrated characterization of the Ni/RGO-1 obtained by reduction of 6 mmol dm^{-3} Ni^{2+} with hydrazine at 353 K for 1 h under N_2 atmosphere in the presence of EDA-GO. (a) SEM image, (b) TEM image, (c) HRTEM image, (d) XRD pattern, and (e) EDX spectrum. (f) The behavior of Ni/RGO-1 under an external magnetic field.